

Spin coherence of holes in GaAs/AlGaAs quantum wells

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The carrier spin coherence in a p-doped GaAs/(Al,Ga)As quantum well with a diluted hole gas has been studied by picosecond pump-probe Kerr rotation with an in-plane magnetic field. For resonant optical excitation of the positively charged exciton the spin precession shows two types of oscillations. Fast oscillating electron spin beats decay with the radiative lifetime of the charged exciton of 50 ps. Long lived spin coherence of the holes with dephasing times up to 650 ps. The spin dephasing time as well as the in-plane hole g factor show strong temperature dependence, underlining the importance of hole localization at cryogenic temperatures.

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Recently the investigation of the coherent spin dynamics in semiconductor quantum wells (QW) and quantum dots has attracted much attention, due to the possible use of the spin degree of freedom in novel fields of solid state research such as spin-based electronics or quantum information processing [1, 2, 3]. Until now the interest has been mostly focused on the spin coherence of electrons, while experimental information about the spin coherence of holes is limited [4]. The hole as a Luttinger spinor has properties strongly different from the electron spin, such as a strong spin-orbit coupling, a strong directional anisotropy, etc. It plays an important role also in coherent control of electron spins, since in many optical schemes charged electron-hole complexes are proposed as intermediate manipulation states [5].

Earlier, the hole spin dynamics in GaAs-based QWs has been measured by optical orientation detecting photoluminescence (PL) either time-integrated or time-resolved [4, 6, 7, 8, 9]. Experimental studies addressed the longitudinal spin relaxation time T_1 [6, 7, 8] and the dephasing time T_2^* , exploiting the observation of hole spin quantum beats [4]. The reported relaxation times vary from 4 ps [6] up to ~ 1 ns [4, 8] demonstrating strong dependence on doping level, doping density and excitation energy. A major drawback of PL techniques is, however, that the spin coherence can be traced only as long as both electrons and holes are present and photoluminescence can occur. Further, they work only for studying the spin dynamics of minority carriers in a sea of majority carriers and are therefore restricted to undoped or n-type doped QWs. However, then the holes can interact with electrons, providing additional relaxation channels via exchange or shake-up processes [8, 10]. These mechanisms can be excluded for p-doped structures if the hole spin relaxation occurs on time scales longer than the radiative annihilation of electrons. A pump-probe Kerr rotation (KR) technique using resonant excitation allows to realize such measurements, which up to now have been reported only for bulk p-type GaN [11] and not yet for low-dimensional systems.

The theoretical analysis of the hole spin dynamics in

QWs has been focused on free holes [10, 12, 13, 14] by considering different relaxation mechanisms: (i) a Dyakonov-Perel like mechanism, (ii) an acoustic phonon assisted spin-flip due to spin mixing of valence band states, (iii) an exchange induced spin-flip due to scattering on electrons, which resembles the Bir-Aronov-Pikus mechanism, but for holes. Recently the attention has been drawn on the role of hole localization and the dephasing caused by fluctuations of the in-plane g factor has been calculated [15].

In this paper we use time-resolved pump-probe Kerr rotation [16] to investigate the spin coherence of holes in a p-doped GaAs/Al_{0.34}Ga_{0.66}As single QW with a low hole density. We find spin dephasing times reaching almost the ns-range at a temperature $T = 1.6$ K with a hole in-plane g factor of about 0.01. Both quantities decrease strongly with increasing temperature, suggesting the important influence of hole localization. We discuss also a mechanism that provides generation of spin coherence for the hole gas under resonant excitation of the positively charged exciton.

The structure was fabricated by molecular-beam epitaxy on a (100) oriented GaAs substrate. A 15 nm-wide GaAs QW was grown on top of a 380 nm-thick Al_{0.34}Ga_{0.66}As barrier and overgrown by a 190 nm-thick Al_{0.34}Ga_{0.66}As layer. 21 nm-thick layers with Al_{0.34}Ga_{0.66}As effective composition realized by GaAs/AlAs short-period superlattices were deposited on both sides of the QW in order to improve interface planarity. Two δ -doped layers with Carbon acceptors were positioned symmetrically at 110 nm distance from the QW. The hole gas concentration and mobility in the QW are $1.51 \times 10^{11} \text{ cm}^{-2}$ and $1.2 \times 10^5 \text{ cm}^2/\text{Vs}$, respectively, as determined by Hall measurements at $T = 4.2$ K. It was possible to deplete the hole density in the QW by above barrier illumination and even invert the majority carrier type, resulting in a diluted electron gas. The sample temperature was varied from 1.6 to 6 K.

A mode-locked Ti:Sapphire laser with a repetition rate of 75.6 MHz and a pulse duration of ~ 1.5 ps (~ 1 meV full width at half maximum) was used for optical excitation.

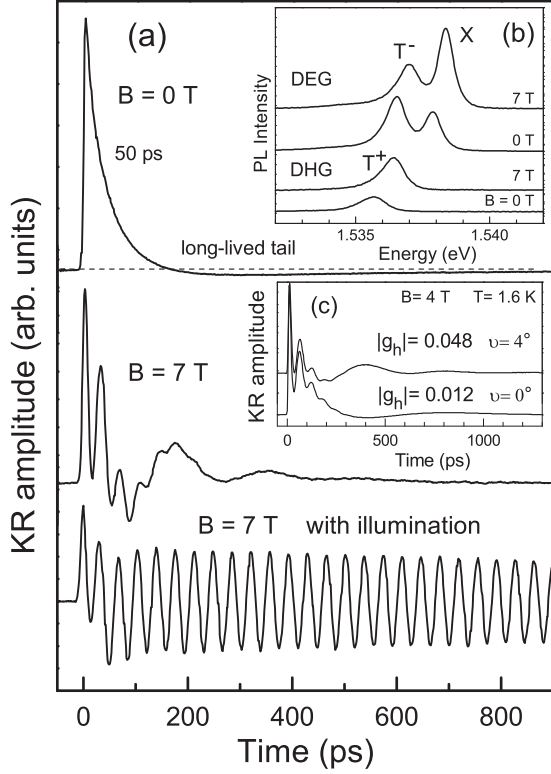


FIG. 1: (a) KR traces for a p-type 15 nm-wide GaAs/Al_{0.34}Ga_{0.66}As QW vs time delay between pump and probe pulses at $B = 0$ and 7 T with field tilted by $\vartheta = 4^\circ$ out of QW plane. Laser at 1.5365 eV is resonant with T^+ line. Power was set to 5 and 1 W/cm² for pump and probe, respectively. Bottom trace was recorded with additional laser illumination at 2.33 eV. $T = 1.6$ K. (b) PL spectra for DHG (excitation at 1.579 eV) and DEG regime (above barrier excitation at 2.33 eV). (c) Comparison of KR traces for $\vartheta = 0$ and 4° .

The laser beam was split into a circularly polarized pump and a linearly polarized probe beam. Both beams were focused on the sample surface to a spot diameter of ~ 100 μm . Magnetic fields $B \leq 10$ T were applied about perpendicular to the structure growth z -axis (Voigt geometry). In a pump-probe KR experiment the pump pulse coherently excites carriers with spins polarized along the z axis. The subsequent coherent evolution of the spins in form of a precession about the magnetic field is tested by the probe pulse polarization. To detect the change of the linear probe polarization plane (the KR angle), a homodyne technique based on phase-sensitive balanced detection was used.

Photoluminescence spectra excited above and below the band gap of the Al_{0.34}Ga_{0.66}As barriers are shown in Fig. 1(b) at $B = 0$ and 7 T. A single PL line corresponding to the positively charged trion T^+ (consisting of two holes and one electron) is seen for the regime of diluted hole gas (DHG) established for below-barrier excitation. After inverting the type of majority carriers to the DEG

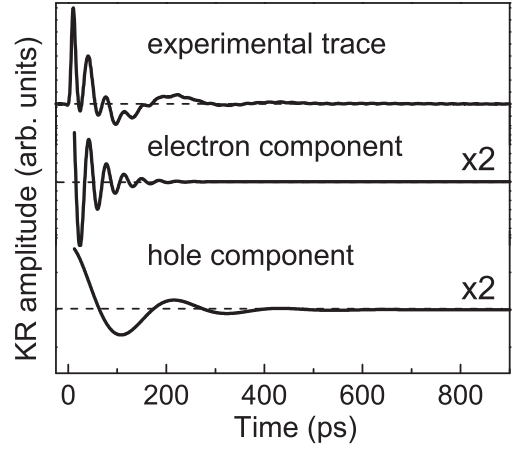


FIG. 2: Top trace: KR signal measured at $B = 7$ T for $\vartheta = 4^\circ$. Bottom traces are obtained by separating electron and hole contributions (see text). Excitation conditions as in Fig. 1.

regime by above barrier illumination the PL spectra consist of the exciton (X) and negatively charged trion (T^-) lines.

The type of majority carriers in the QW can be identified by the KR signals measured at $B = 7$ T, with the laser energy tuned to the trion resonance. The bottom trace in Fig. 1(a) was measured with additional above-barrier illumination (DEG regime) and shows long-lived electron spin beats with a dephasing time of 2.5 ns which is considerably longer than the radiative decay time of resonantly excited triions of about 50 ps. The precession frequency corresponds to a g factor $|g_e| = 0.285 \pm 0.005$, which is typical for electrons in GaAs-based QWs.

Without above-barrier illumination in the DHG regime, fast electron precession is observed only during ~ 200 ps after pump pulse arrival [see middle trace in Fig. 1(a)]. This signal is caused by the coherent precession of the electron in T^+ and disappears with the trion recombination. The electron beats are superimposed on the hole beats with a much longer precession period. The hole beats decay with a time constant of about 100 ps and can be followed up to 500 ps delay. At these long times the KR signal is solely given by coherent hole precession.

Experimentally, it is difficult to observe the hole spin quantum beats due to the very small in-plane hole g factor. To enhance the visibility we tilted the magnetic field slightly out of the plane by an angle $\vartheta = 4^\circ$ to increase the hole g factor by mixing the in-plane component ($g_{h,\perp}$) with the one parallel to the QW growth axis ($g_{h,\parallel}$), which typically is much larger: $g_h(\vartheta) = \sqrt{g_{h,\parallel}^2 \sin^2 \vartheta + g_{h,\perp}^2 \cos^2 \vartheta}$ [17]. The strong change of the hole beat frequency with the tilt angle is seen in Fig. 1(c). The precession frequency is analyzed by $\omega_h = \mu_B |g_h| B/\hbar$, where μ_B is the Bohr magneton, and gives $|g_{h,\perp}| = 0.012 \pm 0.005$ for $\vartheta = 0^\circ$ and $|g_h| = 0.048 \pm 0.005$ for $\vartheta = 4^\circ$.

The electron and hole contributions to the KR ampli-

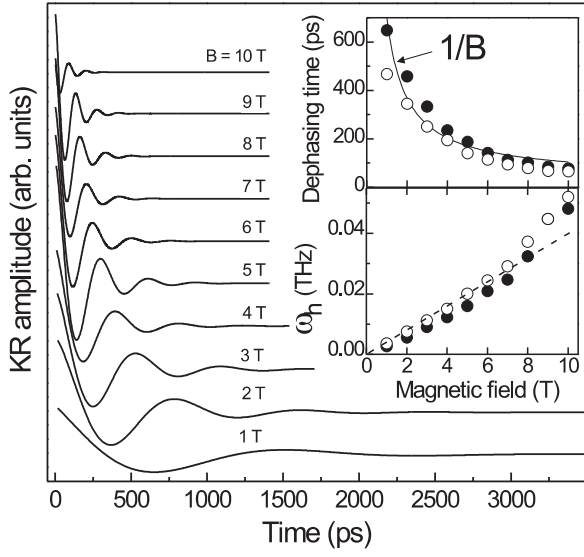


FIG. 3: Hole component of KR signal at different magnetic fields and $\vartheta = 4^\circ$. Top inset: Magnetic field dependence of the hole dephasing time T_2^* . Solid line is $1/B$ fit to data. Bottom inset: Hole spin precession frequency vs magnetic fields. Line is guide to the eye. In inserts closed and open circles show the data measured for pump to probe powers of 1 to 5 W/cm² and 5 to 1 W/cm², respectively. $T = 1.6$ K.

tude, Θ_K , can be separated by fitting the experimental data with a superposition form of exponentially damped harmonic functions for electrons and holes:

$$\Theta_K = \sum_{i=e,h} A_i \exp\left(-\frac{\Delta t}{T_{2,i}^*}\right) \cos(\omega_i \Delta t). \quad (1)$$

A_i are the corresponding signal amplitudes at pump-to-probe delay $\Delta t = 0$, and $T_{2,i}^*$ are the spin dephasing times. An example for a decomposition of the KR signal in the DHG regime is shown in Fig. 2.

Let us turn now to the hole coherence. Figure 3 shows the hole contribution to the KR signal for different B at $T = 1.6$ K. From the fit by Eq.(1) we have obtained the dephasing time T_2^* , which is plotted versus B in the inset. A very long lived hole spin coherence with $T_2^* = 650$ ps is found at $B = 1$ T. With increasing B up to 10 T it shortens to 70 ps. The field dependence can be well described by a $1/B$ -form (see the line in the inset), from which we conclude that the dephasing shortening arises from the inhomogeneity of the hole g factor $\Delta g_h = 0.0007$ in the QW, which is translated into a spread of the precession frequency: $\Delta\omega_h = \Delta g_h \mu_B B / \hbar$. Since $T_2^* \propto 1/\Delta\omega_h$, this explains the $1/B$ -dependence of the dephasing time. The magnetic field dependence of the hole precession frequency in the lower inset of Fig. 3 shows an approximate B -linear dependence up to 7 T. For higher fields a super linear increase is seen which indicates a change of the hole g factor due to mixing between heavy and light hole states, induced by the field.

Two sets of experimental data measured for pump to

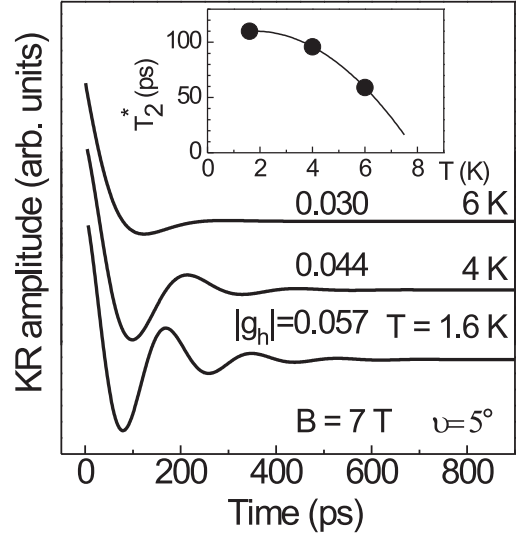


FIG. 4: Temperature dependence of the hole KR signal at $B = 7$ T and $\vartheta = 5^\circ$. Pump and probe powers are set to 1 and 5 W/cm², respectively. Inset: Hole spin dephasing time T_2^* vs temperature.

probe powers 1 to 5 W/cm² and 5 to 1 W/cm² are compared in the insets of Fig. 3. The very similar results demonstrate performance of the experiment in the linear regime for both pump and probe beams with power not exceeding 5 W/cm².

Insight into the origin of the long hole spin coherence can be taken from KR at varying temperatures. The data in Fig. 4 measured at $\vartheta = 5^\circ$ show that (i) the dephasing time T_2^* decreases from 110 down to 60 ps when increasing the temperature from 1.6 to 6 K (see also inset), and (ii) simultaneously the precession frequency decreases notably corresponding to a g factor decrease from 0.057 to 0.030.

These results can be naturally explained by hole localization in the QW potential relief due to monolayer well width fluctuations. The localization energy does not exceed 0.5 meV, which is comparable with the thermal energy at $T = 6$ K. Free holes are expected to have a short spin coherence time T_2 limited by the efficient spin relaxation mechanisms due to the spin-orbit interaction [10, 13, 14]. For localized holes these mechanisms are mostly switched off and one can expect long T_2 times. However, in a KR experiment we do not measure the T_2 time, but rather the ensemble dephasing time T_2^* (Ref. 15), as confirmed by the $1/B$ dependence in the inset of Fig. 3. The T_2^* time gives a lower boundary for the spin coherence time T_2 . Therefore we can conclude, that the T_2 for localized holes is at least 650 ps. Thermal delocalization of holes on the one hand decreases the role of inhomogeneities and reduces Δg_h , which should lead to longer dephasing times. On the other hand, then the fast decoherence of free holes becomes the limiting factor for the spin beats dynamics.

Mixing of heavy and light hole states in a QW is en-

hanced by localization effects. This should be detectable by an increase of the in-plane hole g factor, which is close to zero for free holes [4, 15, 18]. The decrease of the hole g factor with increasing temperature shown in Fig. 4 is consistent with a hole delocalization scenario.

We turn now to discussing the mechanism for optical generation of hole spin coherence in a QW with a DHG. The generation mechanism is similar to the one suggested for singly charged quantum dots [19, 20] and QWs with a DEG [21]. In our experiment pump and probe are resonant in energy with the positively charged trion T^+ . Due to the considerable heavy-light hole splitting, the circularly polarized pump creates holes and electrons with well-defined spin projections, $J_{h,z} = \pm 3/2$ and $S_{e,z} = \pm 1/2$, respectively, according to the optical selection rules [12]. Therefore, $|\uparrow\downarrow\downarrow\rangle$ ($|\uparrow\downarrow\uparrow\rangle$) trions can be generated by a σ^+ (σ^-) polarized pump. Here the thick and thin arrows give the spin states of holes and electrons, respectively.

The pump pulse duration is much shorter than the spin coherence and the electron-hole recombination times. If in addition the pump duration is shorter than the charge coherence time of the trion state the pulse creates a coherent superposition of a resident hole from the DHG and a hole singlet trion T^+ . The spin state of the resident hole with arbitrary spin orientation before excitation can be described by $\alpha|\uparrow\rangle + \beta|\downarrow\rangle$, where $|\alpha|^2 + |\beta|^2 = 1$. Without magnetic field and for fields oriented normal to the z -axis, the net spin polarization of the hole ensemble is zero, so that the ensemble averaged coefficients are equal: $\bar{\alpha} = \bar{\beta}$.

For σ^+ polarized excitation, for which injection of an $|\uparrow\downarrow\rangle$ electron-hole pair is possible, the excited superposition is given by $\alpha|\uparrow\rangle + \beta\cos(\Theta/2)|\downarrow\rangle + i\beta\sin(\Theta/2)|\uparrow\downarrow\rangle$. Here $\Theta = \int \mathbf{d} \cdot \mathbf{E}(t)dt/\hbar$ is the dimensionless pulse area with the pump laser electric field $\mathbf{E}(t)$ and the dipole transition matrix element \mathbf{d} . In general, the hole-trion superposition state may be driven coherently by varying the pulse area, giving rise to Rabi-oscillations as reported recently for (In,Ga)As quantum dots [20]. Such oscillations have not been found yet in QWs, most probably due to the fast carrier dephasing, in particular for strong excitation. Dephasing of the superposition occurs shortly after the pulse on a time scale of a few ps, converting the coherent polarization into a population consisting of holes with original spins \uparrow and \downarrow and trions with $\uparrow\downarrow$.

In a simplified picture, the spin coherence generation can be described as follows: The σ^+ polarized pump creates with certain efficiency trions T^+ of spin configuration $|\uparrow\downarrow\downarrow\rangle$. By this process $|\downarrow\rangle$ holes are pumped out of the DHG, leaving behind holes with opposite spin $|\uparrow\rangle$. Right after the pump pulse the KR signal is contributed by the $|\uparrow\rangle$ hole from the DHG and $|\downarrow\rangle$ electron of the T^+ . The further evolution of the coherent signal depends on

the strength of external magnetic field applied perpendicular to the z -axis.

At $B=0$, the carrier spins experience no Larmor precession. The electron spin relaxation time usually exceeds the lifetime of trions, which is limited by radiative decay, by one-two orders of magnitude. Trion recombination returns the hole to the DHG with the same spin orientation as it was pumped out, if no electron spin scattering occurred in the meantime. This compensates the induced spin polarization and nullifies the KR signal at delays exceeding the trion lifetime. Indeed, the KR signal in the top trace in Fig. 1(a) shows a fast decay with a time constant of ~ 50 ps, which is characteristic for radiative trion recombination in GaAs/(Al,Ga)As QWs [22]. The long-lived tail of the signal has a very small amplitude and is due to hole coherence provided by weak spin relaxation of electrons in T^+ and/or hole relaxation in the DHG during the trion lifetime.

In finite magnetic fields, the carrier spins start to precess about B . Due to the electron spin precession in T^+ , the hole spin returned to the DHG after trion recombination will not compensate the spin polarization of the resident holes. Therefore, a long-lived hole coherence with considerable amplitude will be induced. This coherence is observed in the KR signal as spin beats with low frequency (see Figs. 1 and 3). Note that the Larmor precession of the resident holes may also contribute to generation of hole spin coherence, but the effect is proportional to the ratio of the hole and electron Larmor frequencies and therefore will be rather small.

Let us compare the spin coherence generation for QWs with DHG and DEG resonantly excited in the T^+ and T^- states, respectively. We are interested in a long-lived spin coherence which goes beyond the trion lifetime, i.e. in spin coherence induced for the resident carriers. In both cases the amplitude of the KR signal is controlled by the ratio of the electron spin beat period to the trion lifetime. Nevertheless, the two cases are quite different as for DHG the precessing electron is bound in the T^+ trion, while for DEG the background electron precesses. In the latter case the electron precession in T^- is blocked due to the singlet spin character of the trion ground state.

To conclude, a long-lived spin coherence has been found for localized holes in a GaAs/(Al,Ga)As QW with a diluted hole gas. The spin coherence time exceeds 650 ps and is still masked by the spin dephasing due to g factor inhomogeneities. Localization of holes suppresses most spin relaxation mechanisms inherent for free carriers. It is also worth to note, that due to the p-type Bloch wave functions the holes do not interact with the nuclear spins, which provides the most efficient spin relaxation mechanism for localized electrons [23].

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